Early Validation Analyses of Atmospheric Profiles from EOS MLS on the Aura Satellite

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***Abstract*— We present results of early validation studies using retrieved atmospheric profiles from the Earth Observing System (EOS) Microwave Limb Sounder (MLS) instrument on the Aura satellite. 'Global' results are presented for MLS measurements of atmospheric temperature, ozone, water vapor, hydrogen chloride, nitrous oxide, nitric acid, and carbon monoxide, with a focus on the January through March 2005 time period. These global comparisons are made using long-standing global satellite and meteorological datasets, as well as some measurements from more recently-launched satellites. Comparisons of MLS data with measurements from the Ft. Sumner, New Mexico, September 2004 balloon flights are also presented. Overall good agreeement is obtained, often within 5 to 10%, but we point out certain issues to resolve and some larger systematic differences; some artifacts in the first publicly released MLS (version 1.5) dataset are noted. We comment briefly on future plans for validation and software improvements.**

***Index Terms*— Data Validation, Atmospheric Retrievals**

I. INTRODUCTION

The Microwave Limb Sounder (MLS) is one of four instruments on the Earth Observing System (EOS) Aura satellite. Aura was launched on 15 July 2004 and placed into a near-polar Earth orbit at $\sim$705 km altitude, with a $\sim$1:45 p.m. ascending node time; the main mission objectives are to study the Earth's ozone, air quality, and climate (see [1], [2]). EOS MLS ([3], [4]) contributes to this objective by measuring atmospheric temperature profiles from the troposphere to the thermosphere, and more than a dozen atmospheric constituent profiles, as well as cloud ice water content (see [5]). Other papers in this special issue discuss the MLS retrievals and forward model ([6], [7], [8], [5]), as well as the instrument and its calibration ([9], [10], [11]); see also [12] for a discussion of the MLS data processing system.

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In this paper, we present early validation results from the first publicly-available MLS dataset, version v1.5, with a focus on temperature, ozone (O_3), water vapor (H_2O), nitrous oxide (N_2O), hydrogen chloride (HCl), nitric acid (HNO_3), and carbon monoxide (CO). The early results focus on the stratosphere, although some results are shown for the mesosphere and above, and there is a limited discussion of tropospheric comparisons for temperature and water vapor; useful MLS upper tropospheric retrievals are available in this dataset for ozone and carbon monoxide, but the validation thereof will take more time. For global (or near-global) comparisons, we mainly use the time period from January through March 2005, because MLS version 1.5 data processing started in January 2005, and not much reprocessing for 2004 had been completed at the time of these analyses.

A. Global Correlative Data

For global (or near-global) comparisons, we use datasets from various satellite instruments listed below. Rapid comparisons of MLS measurements with these datasets soon after the Aura launch were enabled by the timely data access provided by the teams who routinely produce these atmospheric products.

- The GMAO GEOS-4 dataset stands for the Global Modeling and Assimilation Office Goddard EOS Data Assimilation System (DAS) output files of meteorological products, delivered to and distributed by the Goddard Distributed Active Archive Center (DAAC). This dataset has been used for comparisons with MLS retrieved temperatures and upper tropospheric humidity (UTH).
- The Challenging Minisatellite Payload (CHAMP) is a small European satellite equipped with a JPL Global Positioning System (GPS) flight receiver and two other antennae, whose occultation-type data, using radio transmitters from several other orbiting microsatellites, are used to make measurements of atmospheric temperature, pressure, and moisture, as well as ocean height and sea-surface winds.
- The Stratospheric Aerosol and Gas Experiment II (SAGE II) is a seven channel solar photometer using ultraviolet (UV) and visible channels between 0.38 and 1.0 microns in solar occultation mode to retrieve atmospheric profiles of O_3, H_2O, and NO_2, along with

aerosol extinction (see [13]). These measurements have been on-going since late 1984, although some degradation in spatial coverage has occurred in recent years, with observations now occurring for roughly half the typical 15 sunrise and 15 sunset events per day. The vertical resolution is typically ~1 km or better. The latitude coverage of SAGE II profiles for the comparisons with MLS discussed here is from 73° S to 66° N; less than 15 % of the hundreds of 2005 January–March profiles used here in comparison to 'coincident' or 'matched' MLS profiles (see section 'C' below) occur in tropical latitudes (20° S–20° N). Version 6.20 SAGE II data files are used.

- HALOE is the Halogen Occultation Experiment, launched aboard the Upper Atmosphere Research Satellite (UARS) in 1991 (see [14]). HALOE measures profiles by solar occultation in the infrared, the products being temperature, aerosol infrared extinction and aerosol properties, and 7 atmospheric constituents, including profiles of O_3, HCl, and H_2O, discussed in this paper. Vertical resolution is about 3-4 km for temperature, 2 km for O_3 and H_2O, and 4 km for HCl. Total uncertainty estimates for these 3 products are typically about 15 to 25 % in the lower stratosphere, and somewhat less in the upper stratosphere, at least for O_3 and HCl *(Dr. Ellis Remsberg, private communication, 2004)*. Latitudinal coverage for the MLS/HALOE comparisons is 75° S to 52° N, with some small gaps. Dates are January 1 through March 13, with a few temporal gaps of a week or more, when HALOE cannot acquire any science data. Over 300 matched profiles are used in the HALOE/MLS comparisons, with only about 10 % of these occurring in the 20° S–20° N range. Version 19 (V19) HALOE data are used here.
- The Polar Ozone and Aerosol Measurement III (POAM III) experiment, launched in 1998, uses solar occultation in the UV/visible to measure O_3, H_2O, NO_2, and aerosols at high latitudes (see [15]). Latitudes in the comparisons shown below cover the ranges 63° N–68° N, and 63° S–82° S; close to 400 profiles are matched with the MLS profiles during January–March, with about half in the northern hemisphere (NH) and half in the southern hemisphere (SH). Vertical resolution is about 1 km and version 4 POAM III data are used.
- The Atmospheric Chemistry Experiment (ACE) is the first mission in the Canadian Space Agency's SCISAT program; ACE was launched on 12 August 2003. The ACE Fourier Transform Spectrometer (ACE-FTS) measurements are the focus of the comparisons presented here. ACE-FTS (hereafter referred to simply as ACE) soundings of the atmosphere are by solar occultation in the infrared (2 to 13 microns) at high spectral resolution (0.02 cm-1). A set of atmospheric profiles for about 20 molecules (as well as temperature) has been derived from these measurements, with a vertical resolution of ~4 km (see [16] for an overview, and [17] for a description of the retrievals). Over 600 version 2.1 profiles, interpolated onto a 1 km vertical grid, are used in the matched comparisons versus MLS, from 1 January to 24 March 24, 2005. For the vast majority (over 99 %) of the comparisons between ACE and MLS shown here, the sampled latitudes are between 41° S–83° S and 56° N–81° N; about 200 profiles are in the SH.
- The Atmospheric Infrared Sounder (AIRS) experiment flies on the Aqua spacecraft 15 minutes ahead of Aura as part of NASA's 'A–Train' ([18]). This ensures that AIRS and MLS observations are closely located in space and time. The AIRS retrieval system uses a combination of infrared and microwave nadir observations to infer profiles of temperature and water vapor, along with cloud and surface properties ([19]). AIRS retrievals have a 45 km horizontal spacing on a swath that is ~ 1500 km wide. AIRS profile uncertainties in the troposphere are ~1 K for temperature in 1 km layers, and 15 % for humidity in 2 km layers ([20]). Temperature resolution drops to 2 K in 2 km layers in the stratosphere; AIRS has little sensitivity to H_2O for mixing ratios less than about 10 ppmv ([21]).

B. Ft. Sumner 2004 Balloon Campaign Data

For the 2004 Fall balloon campaign from Ft. Sumner, New Mexico, we consider the Observations of the Middle Stratosphere (OMS) *in situ* gondola data from the flight of 17 September, and a separate flight of *in situ* measurements on 29 September 2004; these datasets come from the following instruments:

- The ozone photometer is a dual channel UV photometer measuring ozone *in situ* from the surface to the maximum balloon altitude with 1 second resolution (5 m vertical) and an accuracy of 3–5 % (for a recent description, see [22]).
- ALIAS–II is the balloon-borne version [23] of the Airborne Laser Infrared Absorption Spectrometer. It is a two-channel tunable laser spectrometer configured to measure *in situ* HCl using an interband cascade laser at 3.3 microns, and CO using a quantum cascade laser at 4.6 microns. ALIAS–II has flown several times before, and uses an open-path Herriott cell (path 64 m) extending out from the gondola. Calibration for CO is done preflight using certified gas mixtures. Calibration for HCl is done in-flight by scaling to several nearby ozone lines using the measurements of the ozone photometer on the same gondola. For the 17 September 2004 flight, absolute uncertainty in the measured HCl is about 10 % or 0.1 ppbv, whichever is larger. CO has been measured numerous times by the aircraft instrument ALIAS [24], but here with ALIAS–II for the first time on a balloon platform. Numerous spikes of ~500 ppbv, removed from the dataset, were seen on ascent and descent, and are tentatively attributed to contamination, possibly from liquid nitrogen boil-off. This contamination increases the absolute uncertainty for measured CO to ~10 % for tropospheric values, and to ~30 % for stratospheric values.
- LACE is the Lightweight Airborne Chromatograph Experiment, which can provide accurate *in situ* profiles of

various halons, chlorofluorocarbons, as well as methane, N_2O and CO. Its 17 September 2004 measurements of N_2O are used here for comparison to MLS N_2O profiles. The total uncertainty in LACE measurements is small (of order 1 to 2 %) in comparison to the precision of individual MLS retrievals; see [25] for further details about this instrument.
- Also, a cryogenic whole air sampler (CWAS) collected samples of various gases above Ft. Sumner on 29 September 2004. The CWAS is a new version of a liquid-neon-based sampler described originally by [26]. Twenty five samples were collected. The N_2O measurements reported here were analyzed by gas chromatography with electron capture detection using the technique given in [27]; total uncertainty in these measurements is estimated to be of order 1 %.

The second Ft. Sumner 2004 balloon dataset used here for comparisons is from the 23–24 September flight of remote sounding instruments obtained from the Balloon Observations of the Stratosphere (BOS) gondola. This includes measurements made near the time of the daytime Aura overpass, as well as nighttime data early on September 24, when nighttime overpass data from MLS provide the more appropriate comparison points. Available measurements from the following BOS instruments are considered here:

- The JPL MkIV instrument is a solar occultation Fourier Transform Infrared (FTIR) spectrometer that measures the entire 650 to 5650 cm-1 region simultaneously at 0.01 cm-1 spectral resolution ([28]). Profile information is obtained at sunset or sunrise. Although MkIV profiles are retrieved on a 1 km vertical grid, their true vertical resolution is 2–3 km. The MkIV error bars shown in this paper (and taken from the data file) represent the 1–sigma measurement precisions.Systematic errors in the stratospheric profiles caused by spectroscopic uncertainties could be as large as 6 %, 7 %, 5 %, 5 %, 12 %, and 5 % for the MkIV retrievals of O_3, H_2O, HCl, N_2O, HNO_3, and CO, respectively.
- The Smithsonian Astrophysical Observatory (SAO) far-infrared spectrometer (FIRS)-2 is also an FTIR spectrometer but it measures atmospheric thermal emission at long wavelengths between 6 and 120 microns, with a spectral resolution of 0.004 cm-1 ([29]). As for MkIV, many stratospheric profiles are obtained from these measurements. Vertical resolution is similar to that of MkIV; plots shown here use error bars based on the estimated uncertainties from random spectral noise and errors in atmospheric temperature and limb pointing angle.

We also point out that important OH and HO_2 data from the EOS MLS instrument are being validated by using measurements from the Ft. Sumner flight, using FIRS-2 OH and HO_2 data as well as the Balloon OH (BOH) data; this work will be described elsewhere.

C. Comparison Approach and Methods

For most MLS products discussed here, we do not attempt to extend the analyses to the highest and lowest altitudes of the retrieval capabilities, given the early stages after launch and the relative paucity of correlative datasets for such detailed comparisons in the troposphere and mesosphere (or higher). For the comparisons presented below (unless otherwise noted), we use 'matched' pairs of profiles from MLS and the correlative dataset(s); the typical criterion used here for a 'match' or 'coincidence' is closest profile within 1° of latitude (North or South), 12° of longitude (East or West), and on the same day; we note some slightly different criteria, if used, in the relevant Figure captions. More detailed sensitivity analyses will be performed later, and with longer time series, but a few comments are provided below for cases that appear to benefit significantly from different coincidence criteria. Also, the issue of how to best account for the vertical resolutions of different measurement systems has been largely ignored in this early set of comparisons. Unless otherwise indicated, simple interpolation of profiles (as a function of log(pressure)) to the fixed MLS pressure grid is carried out. We believe that for the majority of global comparisons, these effects will have negligible impact on the overall biases that are observed, since the vertical resolution of most of the other instruments is not very different from that of MLS; also, comparisons of finer resolution average profiles from SAGE II to the 'MLS-gridded' average profiles reveal little overall change in the level of agreement versus MLS. A more detailed approach has been undertaken for the MLS/AIRS water vapor comparisons in the upper troposphere, as discussed in the H_2O section.

A 'data quality' document, made publicly available to users of MLS data, contains more information about the effect of clouds on MLS retrievals and on data quality, along with recommended methods for rejecting poor quality profiles. The details of this additional screening are expected to evolve as more is learned from future in-depth analyses. However, this should not have a big impact on the overall results shown in this paper, since only a small fraction of profiles is expected to be screened out by more detailed analyses, and most of the impact will be at pressures of 100 hPa or higher.

Unless otherwise stated, the statistics of differences for the plots shown in this paper are given with respect to the correlative dataset (say 'C'), namely averaged MLS profiles minus averaged C profiles; the average difference ('d') for each height, is then expressed as a percent of the average correlative dataset, namely 100 x d/(avg. C). A similar procedure is followed for the standard deviations of the differences; these are calculated in absolute (e.g., mixing ratio) units first, and then expressed as a percent of the average correlative dataset values. Clarification of the procedure is needed so that different groups can intercompare or duplicate results with minimal confusion about such issues. Estimated uncertainties (or 'errors') in the mean differences shown in this paper are calculated by combining the estimated random errors typically found in files of atmospheric products. These errors are then also expressed as a percentage of the mean correlative data. Calculating the errors in the mean differences based, instead, on the scatter in these differences, often results in somewhat larger errors, since they include atmospheric variability and not just measurement noise estimates. However, the errors in the mean differences between MLS and correlative global data shown in this work tend to be small enough that the mean differences are most often statistically significant and not dominated by random errors. We conservatively choose to show twice the errors in the mean for the global plots below.

II. Temperature

The standard MLS temperature product for v1.5 is taken from the 118 GHz ('Core' phase) retrieval at and below 1 hPa and from the 190 GHz ('Core+R2' phase) retrieval at and above 0.68 hPa; see [6] for more information on the various retrieval phases. The retrieval is produced from 316 hPa to 0.001 hPa and should be useful for scientific study over this range, although the bottommost level is somewhat noisy and there is indication of vertical oscillations in the lower stratosphere. Validation is still extremely preliminary above 0.1 hPa. Typical estimated precisions are 2.2 K at 316 hPa, 1 K at 100 hPa, 0.5 K at 10 hPa, 0.8 K at 1 hPa, 1 K at 0.1 hPa, 1.2 K at 0.01 hPa, and 2 K at 0.001 hPa. For this data version, the vertical resolution of the temperature retrieval is $\sim$ 4km in the middle stratosphere but degrades to worse than 12 km in the mesosphere and 10 km in the upper troposphere. Current temperature retrievals using the 240 GHz radiometer ('Core+R3' phase) have significantly better vertical resolution in the troposphere (~ 4 *km*) but exhibit significant vertical oscillations. We expect to realize this better vertical resolution in future data versions.

In Figure 1, we compare retrieved values of MLS temperature to ACE, HALOE, AIRS v3, CHAMP GPS and to the interpolated GMAO GEOS-4 analysis. ACE, HALOE and GEOS-4 coincidences are from January-March, 2005, while AIRS (v3 data) and CHAMP comparisons are from January only. GEOS-4 is used as the MLS retrieval *a priori* temperature, but a large *a priori* error is used where MLS has good sensitivity, limiting the impact of the *a priori* upon retrieved values; a 20 K error is assigned in the stratosphere and 10 K in the upper troposphere. The GEOS-4 analyses are spatially and temporally interpolated to the MLS observation points. The HALOE and CHAMP (GPS occultation) comparisons are for profiles separated by less than 6 hours and 300 km. The ACE coincidence criteria are 1° of latitude and 12° of longitude on the same UT day. AIRS profiles are averaged to a 2.5°x3.5° lat/lon grid and the coincidence criteria are 100 km and 12 minutes.

The CHAMP GPS occultation measurements have an advertised mean bias of less than 0.1 K and typical individual profile accuracies of 0.5 K, approaching 0.2 K at the tropopause ([30], [31]) This accuracy is degraded when contributions of water vapor to the index of refraction are large and uncertain, but this does not present a problem in the dry stratosphere and tropopause region, so CHAMP may provide the least biased comparison set in this region. The comparisons shown suggest that MLS temperatures have a 1-2 K warm bias in the stratosphere. The bias with respect to CHAMP GPS occultation profiles in the lower stratosphere is at the low end of this range, typically slightly less than 1 K. As the sets of coincidence profiles are not the same for the different instruments, the results shown do not necessarily indicate that the correlative datasets are inconsistent with one another, but there is a suggestion of a small cold bias in the AIRS v3 stratospheric temperatures. AIRS v4 temperatures are not expected to differ significantly from v3.

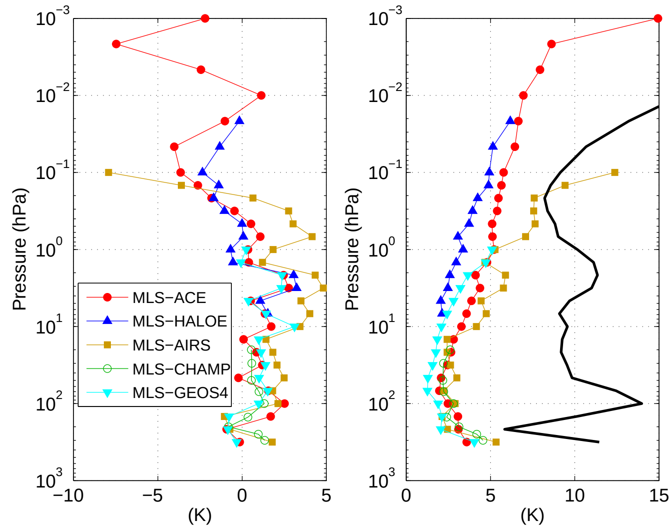

Fig. 1. The left panel shows mean MLS-minus-comparison-set temperature differences averaged over sets of matched ('coincident') profile pairs from the first three months of 2005. The AIRS and CHAMP pairs are only from January, but have 12,438 and 3074 matched profiles with MLS, respectively. The HALOE comparisons include 284 matched profiles, while the ACE comparisons include 525 matches. The GMAO comparisons lead to a large number of matched profiles, namely over 215,000. The right panel shows standard deviations of these differences, for each set of profile pairs. The thick black line is the standard deviation of all MLS profiles in the 3 month period.

The thick black line on the right panel of Figure 1 is the 1-σ variability of the MLS retrievals over all v1.5 retrievals in January-March 2005, and primarily reflects atmospheric variability; MLS noise (less than 1 K for single profiles) is a minor contributor. There is generally good agreement between the variability of MLS and comparison datasets over the sets of profiles compared, as expected; GEOS-4 variability is typically within 0.6 K of the black line below 0.46 hPa. ACE variability and MLS variability over their compared profiles agree to better than 1 K below 0.002 hPa. Where the standard deviations of the differences are small compared to the atmospheric variability, the instruments are capturing the same variance, as they should. The 1-day time coincidence criterion used with ACE may contribute to biases in the mesosphere, as tides can be aliased by persistent differences in the time samplings of the two satellite instruments.

III. Ozone

The standard product for O_3 in version 1.5 is taken from the 240 GHz (Core+R3) retrievals. These data are considered useful for scientific studies from 215 to 0.46 hPa, with some caveats noted below, based on comparisons with retrieved profiles from other satellite measurements as well as from other MLS radiometer bands. Some averaging over time and space is recommended for the upper tropospheric region, where useful O_3 results are being obtained, but this is beyond the scope of validation studies to be presented here. Simulations for the standard O_3 product indicate that this product has the highest sensitivity down into the upper troposphere, as well as in the mesosphere. However, because of larger differences between the 240 GHz band results and other MLS bands in the mesosphere (see below), as well as the difficulty associated with large diurnal effects in this region and their potential impact on occultation profiles, we defer more careful studies of this region to future work. Retrieval simulations indicate that average biases (from the retrieval process itself) are small over the vertical range recommended above, with overall accuracy (closure) of better than ∼1%, not a major error source; an iterative full forward model is used for the standard product retrieval. The estimated single-profile precision reported by the Level 2 software typically varies from ∼0.2 to 0.4 ppmv (or 2 to 15 %) from the mid-stratosphere to the lower mesosphere; the observed scatter in the data, evaluated in a narrow latitude band centered around the equator where atmospheric variability is expected to be small, tends to be slightly larger in most of this region. This scatter is larger than the estimated precision by ∼30% near the ozone peak, and by a factor of more than two near 100 hPa. The vertical resolution of the standard product for O_3 is ∼2.7 km over the range 147 to 0.2 hPa, degrading to ∼4 km at 215 hPa.

A. Global Comparisons

Figure 2 gives a broad comparison of the latitudinal variations in MLS O_3 data, from one early day of measurements (August 30, 2004), to the average of HALOE data from August 2004 and the average of SAGE II data from August and September 2004. Overall agreement for other products like HCl and H_2O (not shown) is similar in nature. The power of day and night global coverage (∼ 3500 profiles every 24 hours) from the MLS emission measurements is demonstrated by such a plot.

To get a more accurate assessment of differences between MLS and other global ozone datasets, we now provide more detailed analyses of matched O_3 profiles during the January through March 2005 time period. Figure 3 gives results of matched comparisons between ozone profiles from MLS and SAGE II, with average profiles, average differences and standard deviations of these differences, as explained in the Introduction. Figures 4, 5, and 6 are the same kind of comparison, but versus HALOE, POAM III, and ACE data in January–March 2005; latitudinal coverage for the various comparisons has been described in the Introduction. For the POAM III comparisons, it was found that reducing the time coincidence criterion from ‘same day’ to (plus or minus) 3 hours led to a significant reduction (by up to a factor of two) in the biases. Overall, these comparisons indicate that MLS ozone values tend to be slightly high in the lower stratosphere, and slightly low in the upper stratosphere, but the degree of ’tilt’ in this slope of the average differences (right panels) changes from one comparison to the next. It is most accentuated in the ACE comparison, and least in the HALOE plot, where 5 % agreement is observed for essentially the whole range from 100 to 1 hPa. SAGE II stratospheric values agree this well with MLS also, except in the region near 1 hPa, where MLS is lower

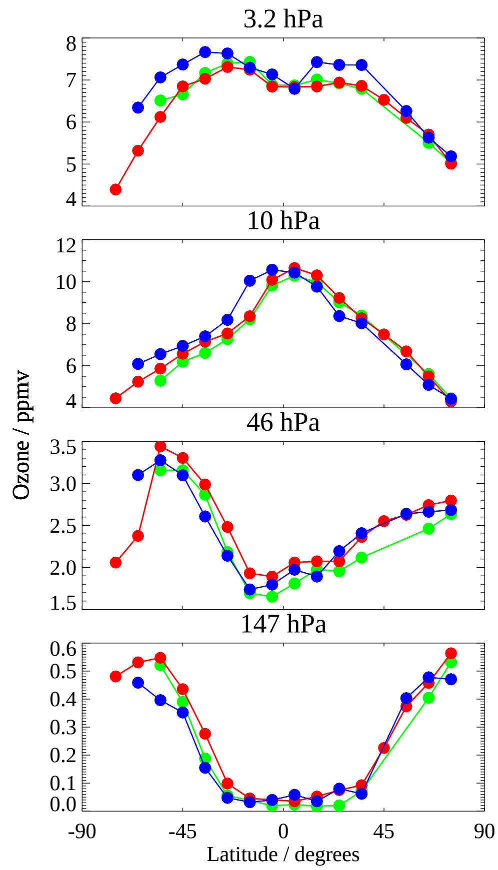

Fig. 2. Early sample comparison between zonal mean MLS (red), SAGEII (blue), and HALOE (green) ozone measurements as a function of latitude at four pressure levels. The MLS measurements are for 30 August 2004, in 10°latitude bins. The SAGE II and HALOE data combine sunrise and sunset occultations in the same latitude bins, using all August occultations for HALOE and all August and September data for SAGE II.

by 10 to 15 %. ACE ozone values in the 40–55 km region have been shown to be on the high side of SAGE III and POAM III measurements by as much as 38 % and 28 % (see [32]). This would seem to explain at least some of the differences seen versus MLS in this region (in Figure 6), where ACE values are also larger than MLS (by ∼10–20 %). The ACE ozone values are also larger (by ∼ 0.4 ppmv) than matched HALOE profile values above ∼35 km, according to the study by [33]. These comparison plots also show that the atmospheric variability is generally very well matched between MLS and the other satellite observations (based on a comparison of the size of the error bars in the left panels of these plots). Moreover, there are many instances where variations near the NH vortex edge or in and out of the vortex are well tracked by both MLS and the other satellite measurements, based on plots of matched profiles on individual days (not shown here). In the year ahead (and leading to a new MLS software version), we intend to pursue issues raised by these comparisons by adding more information as a function of season and latitude, and by investigating potential MLS issues in the mesospheric and upper stratospheric retrievals. Indeed, we see from Figure 7 that the other ozone bands tend to produce smaller values for

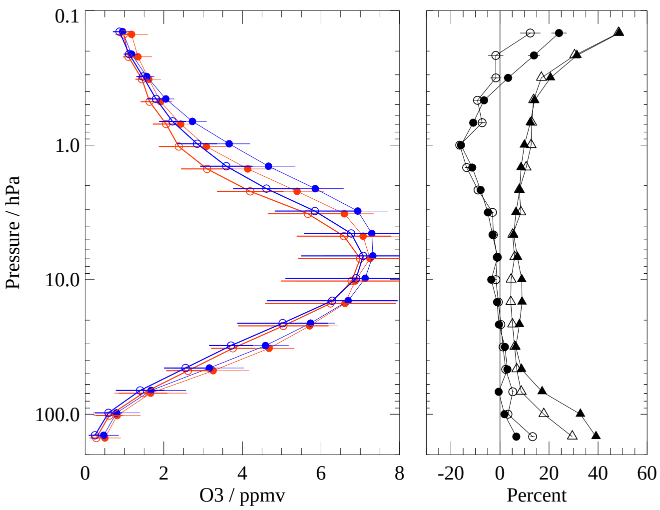

Fig. 3. MLS and SAGE II O_3 comparisons. Left panel: Averages of the matched profiles from MLS (red) and SAGE II (blue), with closed and open circles corresponding to NH and SH profiles. The standard deviation of the profiles is shown by the error bars; a slight shift in pressure between the MLS and other dataset is introduced for clarity, but the SAGE II profiles have been interpolated to the MLS retrieval grid for these plots. Right panel: Average differences (circles) are given for MLS minus SAGE II abundances, expressed as a percent difference from the corresponding average SAGE II profile, with error bars representing twice the estimated error in the means; if no error bar is apparent, it is small and hidden by the symbol itself. Also shown are the standard deviations of the differences (triangles), with closed and open symbols referring to the NH and SH, respectively. A total of 873 profiles was used in these matched comparisons; a match means the use of the closest MLS profile within plus or minus 1°latitude and 12°longitude of each SAGE II profile, and within 24 hours (on same day).

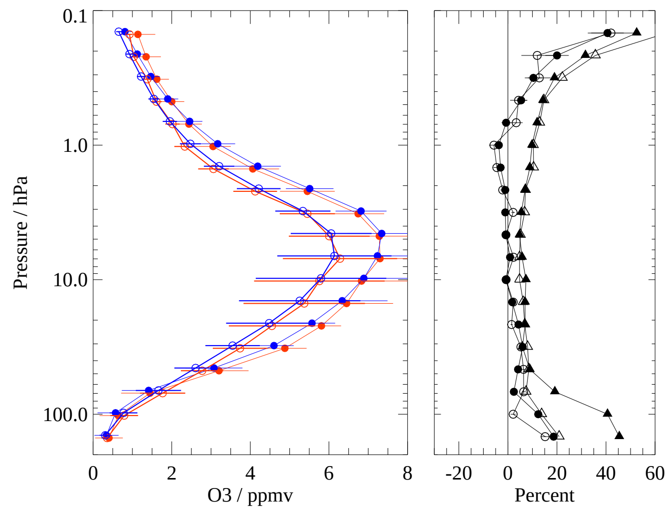

Fig. 4. As Figure 3, but for MLS and HALOE O_3 comparisons; a total of 303 matched profiles was used in this case, with a time coincidence criterion of plus or minus 6 hours.

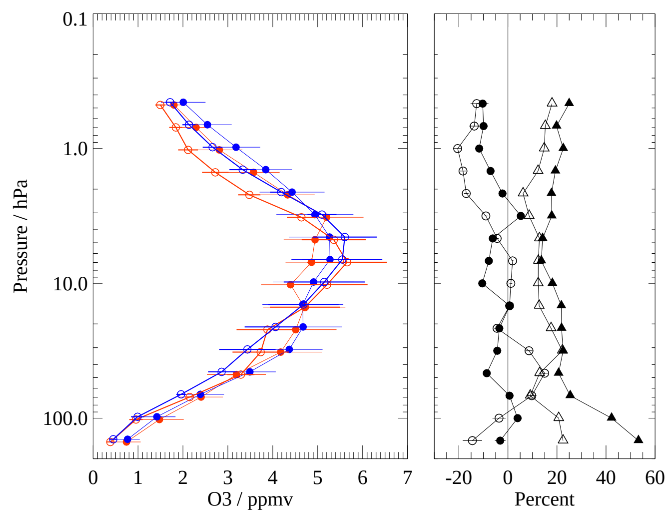

Fig. 5. As Figure 3, but for MLS and POAM III O_3 comparisons, and with a tighter time coincidence criterion of (plus or minus) 3 hours; see text. A total of 407 matched profiles was used in this case.

lower mesospheric O_3 than the standard O_3 product; this may be in part because of issues relating to the narrower spectral channels (digital autocorrelator channels or DACs) used for the 240 GHz band retrievals. Any overestimate in the mesosphere could lead to some overcompensation near 1 hPa, but this remains to be investigated. Otherwise, Figure 7 (for January 2005) and similar plots for other months indicate that a pretty systematic bias exists between the various MLS O_3 bands. We also observe (from plots not shown here) that the stratospheric retrievals for the standard MLS product lead to a better overall match versus the other satellite measurements, and that the tilted nature of some of these differences is not a feature of the standard MLS product alone. The upper stratospheric agreement between bands is often at about the 7 % level or better, which is consistent with a (rough) 5% accuracy estimate for each band; we believe that such an accuracy figure is indeed achievable, with most of the uncertainty in this region arising from spectroscopic uncertainties or inconsistencies between the various bands. More complete error analyses, including the impact of any pointing knowledge uncertainties (currently believed to be a small contributing factor, see [11]) will be pursued later. The larger differences in the lower stratosphere, notably for the 640 GHz band, also need further investigation, in terms of the spectroscopic parameters in this spectral region, as well as continuum effects.

B. Ft. Sumner Comparisons

Figure 8 shows a comparison of MLS ozone with profiles obtained from the Ft. Sumner, New Mexico, balloon flights of September, 2004 (see Introduction section). The two closest day and night MLS profiles are compared to the FIRS-2 profiles closest in time to the Aura overpass, for 23 September and the night of 24 September. The MkIV profile from 23 September (during local sunset) is also shown; this is to be compared to the daytime MLS profiles, since it was taken about 5 hours later than these measurements. We also show in Figure 8 the *in situ* fine resolution data from the ozone photometer instrument aboard the OMS gondola on 17 September 2004; this is to be compared to the appropriate MLS daytime profiles also shown for that day. It is difficult to tell from these measurements whether some of the systematic

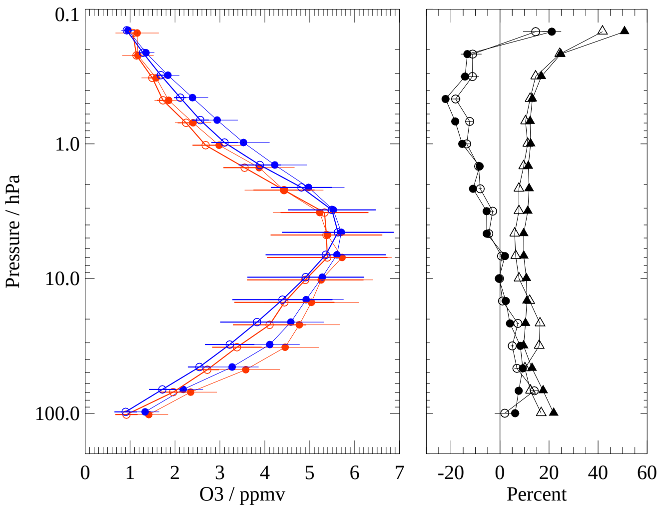

Fig. 6. As Figure 3, but for MLS and ACE O_3 comparisons. A total of 622 matched profiles was used in this case.

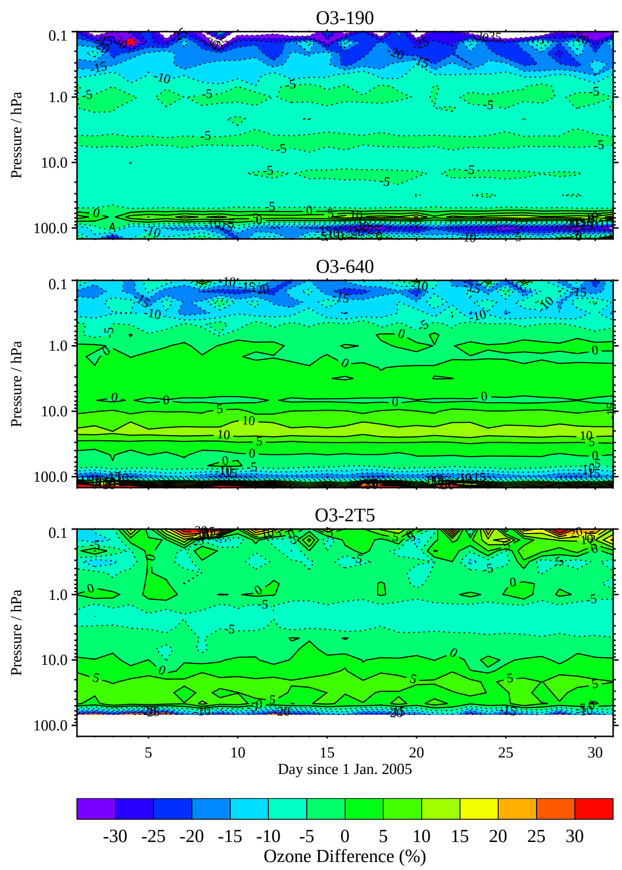

Fig. 7. Percent differences between the various MLS retrievals of O_3 (top panel for 190 GHz, middle panel for 640 GHz and bottom panel for 2.5 THz) and the standard (240 GHz) MLS O_3 retrieval, during January 2005. A positive difference means that the non-standard ozone product gives a larger value than the standard product.

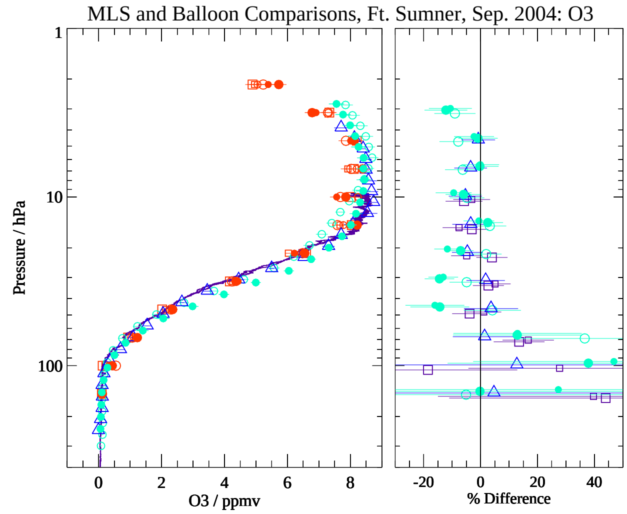

Fig. 8. MLS O_3 compared to Ft. Sumner balloon flight measurements in Sep. 2004. Left Panel: As explained in the text, the two closest MLS profiles for daytime and nighttime are plotted for mid-day on Sep. 23 and the early morning of Sep. 24, respectively. Open red circles are for daytime MLS profiles, with the larger symbol being for the closest profile to the FIRS-2 profile (open cyan circle) that is closest in time to the daytime Aura overpass. Closed red circles are the same but for the nighttime FIRS-2 profile (closed cyan circle) closest in time to the nighttime Aura overpass. Open blue triangles are the profile from MkIV sunset data on the evening of Sep. 23, to be compared to the daytime MLS profiles. Error bars on the MLS and remote balloon measurements are based on estimated errors from each experiment (essentially the random error component). The purple fine resolution profile is from the OMS *in situ* photometer data, for Sep. 17; this is to be compared to the two open red squares, representing the two closest daytime MLS profiles for that day's flight. Right Panel: Percent differences (for MLS minus balloon data) are shown, with symbols referring to the balloon measurements listed in the left panel caption, but with open squares for the *in situ* photometer comparison. Error bars give twice the random errors in these differences.

differences observed in the previous section are duplicated here, given the error bars, although the MLS data are slightly on the low side in the 3 to 10 hPa region, by a few to 10 %, depending on which correlative dataset is being used in the comparison. There are similar differences in places between the MkIV and FIRS-2 profiles, and the several other FIRS-2 profiles, not shown here, also exhibit such variations, some of which could arise from different sampling locations and times, and some from random errors. Given the lack of sufficient regular balloon launches for statistical comparisons, more careful comparisons between MLS and ozonesondes will be useful in order to understand potential issues and interpret the satellite measurements near 100 hPa, where the profile vertical gradient changes steeply; appropriate consideration of the differing vertical resolutions also needs to be investigated for optimum comparisons. Overall, the Ft. Sumner ozone comparisons provide quite good agreement.

IV. Water Vapor

The standard product for H_2O in version 1.5 is taken from the 190 GHz (Core+R2A) retrieval. The recommended range for single profiles for scientific studies extends from 316 hPa to 0.1 hPa. Above 0.1 hPa the poorer signal-to-noise ratio and smaller mixing ratios results in much larger estimated precisions, and these data should be used only after consultation

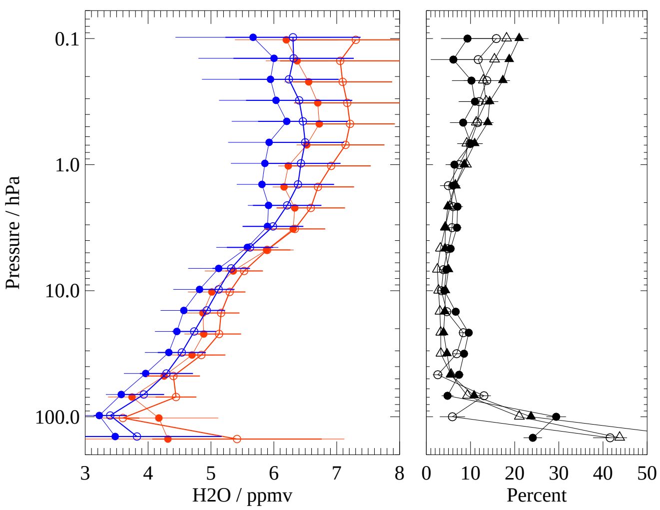

Fig. 9. As Figure 4, but for MLS and HALOE H_2O comparisons. A total of 326 matched profiles was used in this case.

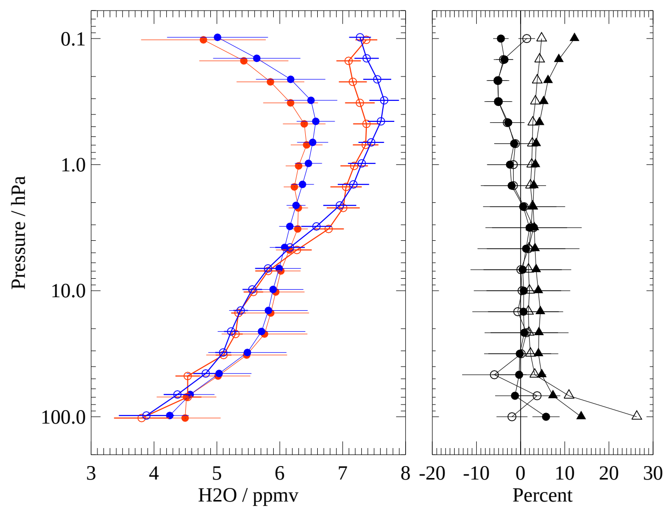

Fig. 10. As Figure 3, but for MLS and ACE H_2O comparisons. A total of 616 matched profiles was used in this case.

with the MLS team. Typical single-profile precisions reported by MLS Level 2 v1.5 software is the larger of 17% or 2 ppmv at 316 hPa, the larger of 10% or 0.9 ppmv at 215 hPa, 0.7 ppmv at 147 hPa, and 0.5 ppmv at 100 hPa. For most of the stratosphere, the estimated precision is ∼0.2-0.3 ppmv; this increases to ∼0.7-0.8 ppmv in the middle mesosphere. Scatter in the real data for regions of small atmospheric variability, and in retrieval simulations, suggests lower measurement precisions, with typical values of ∼0.1 ppmv for most of the stratosphere. Vertical resolution is estimated to be 3 km in the upper troposphere and lower stratosphere, ∼4 km in most of the stratosphere, and ∼6 km in the lower mesosphere.

A. Global Comparisons

1) Stratosphere: In a manner similar to the ozone comparisons, we show in Figure 9 average results for matched MLS and HALOE H_2O profiles during the January through March 13 time period. The mean profiles show the expected differences between NH and SH, with clearly lower mixing ratios for the winter hemisphere in the upper stratosphere and lower mesosphere. On average, MLS H_2O has a positive bias with respect to HALOE H_2O at all pressures. In the middle and upper stratosphere, this bias is ∼5-10%. This is consistent with the findings of the SPARC report [34], which notes that HALOE H_2O values have a ∼5% dry bias with respect to the mean of all H_2O measurements compared. In the middle mesosphere, the MLS bias versus HALOE increases to ∼10-15%. In the lower stratosphere, the bias also grows, and oscillations can be observed in the average of the differences. This can be attributed to some 'zigzags' in the individual MLS profiles at these pressures, related to the difficulties of getting radiance closure in the upper troposphere, a known artifact for this version of the data. Clear differences between NH and SH comparisons are found in the lower stratosphere, where the oscillations in the MLS averages of Figure 9 go in opposite directions.

Figure 10 gives results of matched comparisons between MLS and ACE H_2O profiles over a pressure range similar to the one shown for HALOE. In most of the stratosphere, the agreement is excellent, with very small biases between the two sets of measurements. In the lower stratosphere, the biases increase, partly due to the oscillations in the MLS profiles, as mentioned above. This MLS artifact is more prominent for the SH comparisons, although it appears that the oscillations are centered on values close to the ACE average profile. In the lower mesosphere, there is an increasing negative bias (MLS values lower than ACE values), which nevertheless remains below 5%.

Figure 11 gives results of matched comparisons between MLS and SAGE II H_2O profiles. In the 15 to 40 km region, where the SAGE II retrievals are known to work well, the differences between the instruments are in the 0-20% range, with larger biases in the lower stratosphere. Above ∼40 km, the SAGE II retrievals become increasingly noisy, as can be seen in the variability (error bar) shown in the average profiles. In the upper stratosphere the bias changes sign and becomes negative, as was observed for the ACE comparison, suggesting a possible negative bias for the lower mesosphere MLS retrievals; however, much more work remains to be done to arrive at a firm conclusion on this issue.

Figure 12 gives results of matched comparisons between MLS and POAM III H_2O profiles. In comparison to the previous plots, these results show some distinct differences for the NH and SH, both in the averages and standard deviations of the differences. For both NH and SH averages, MLS has a negative bias in most of the stratosphere, with the larger values in the SH up to ∼-20%. We believe that these differences between NH and SH are caused by a sunrise/sunset bias reported previously for comparisons between SAGE II [35] and HALOE [34] with POAM III. The larger standard deviation for the NH comparisons could be attributed to a larger spatial variability for the winter hemisphere. As the POAM III NH measurements in this period are taken in a

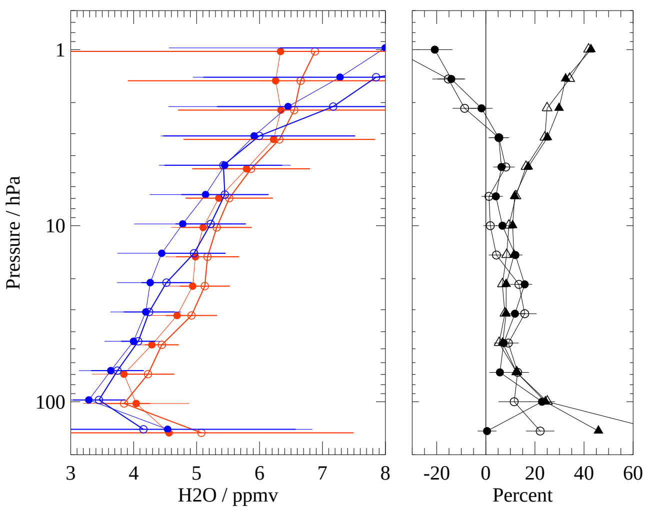

Fig. 11. As Figure 3, but for MLS and SAGE II H_2O comparisons. A total of 273 matched profiles was used in this case, with a time coincidence criterion of plus or minus 6 hours.

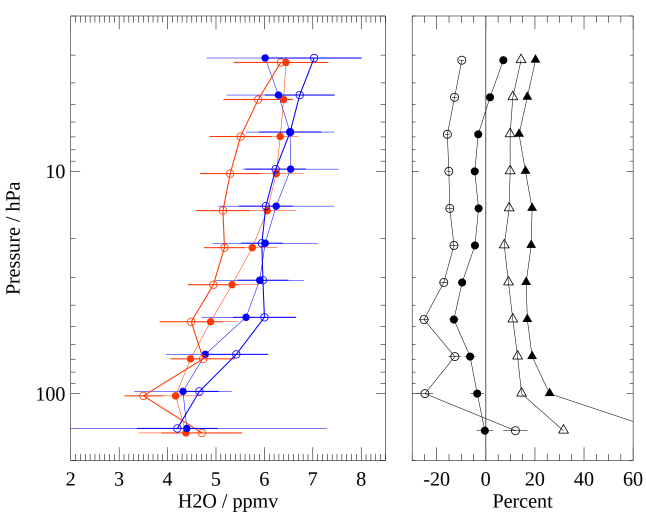

Fig. 12. As Figure 5, but for MLS and POAM III H_2O comparisons.

narrow latitude band around 65° N, the proximity of the winter vortex makes the co-location more challenging in terms of time and distance. Indeed, it was found that tightening the temporal coincidence criterion to 3 hours instead of 'on the same day', helped to reduce the ozone biases between MLS and POAM III by a factor of two at some heights; there is a smaller impact on these comparisons for H_2O.

2) Upper Troposphere: MLS retrievals of H_2O are made into the upper troposphere, as has been shown successfully from the UARS MLS results on this important atmospheric measurement [36]. The EOS MLS measurements in the upper troposphere seem to behave as expected, based on comparisons versus UARS MLS, GMAO GEOS-4, and AIRS data. More detailed validation versus radiosondes and other available datasets (and campaign-mode aircraft data) are in progress.

Here, we give a sample comparison with AIRS V4.0 shown in Fig. 13 for 5 November 2004. The AIRS v4.0 dataset only exists for special focus days produced by the AIRS team. These focus days are produced at a rate of about one per month and, at the time of this writing, there were only four such days for which the EOS MLS data were processed to Level 2. The Aura and Aqua spacecraft fly in formation with Aura ∼15 minutes behind. With EOS MLS looking forward and AIRS nominally looking nadir, apart from differences concerning horizontal and vertical footprints, exact coincidences separated by 8 minutes are available for all the EOS MLS profiles. After reading the appropriate files, the closest coincident AIRS profile is found for each EOS MLS profile. The AIRS profile is quality screened using the `Qual_Temp_Profile_Mid` flag. The AIRS profile is gridded on the standard assimilation levels (1100, 1000, 925, 850, 700, 600, 500, 400, 300, 250, 200, 150, 100, 70, ..., hPa). The AIRS H_2O concentrations are interpreted as a constant mixing ratio between the pressure value assigned to the concentration and the pressure grid point above it. The AIRS profile is interpolated to a fine vertical grid similar to the EOS MLS FOV scan and then fitted (smoothed) to the MLS retrieval grid points using least-squares [7]. The comparison for AIRS v4.0 for one orbit is typical of all comparisons done to date. There are three MLS vertical levels where both instruments overlap well. EOS MLS currently does not retrieve H_2O at pressures higher than 316 hPa and AIRS loses sensitivity to H_2O at values less than 10 ppmv or, typically, at pressures around 150 hPa. The instruments show good tracking; however, there are many AIRS profiles that fail their `Qual_Temp_Profile_Mid` flag and hence show as gaps. Gaps are more likely at high latitudes under dry conditions. Comparisons between AIRS and MLS for two focus days, 5 November 2004 and 23 December 2004, show that the latter is 25% drier, 3% wetter and 12% wetter at 316, 215, and 147 hPa respectively. Comparisons for 18 days with the V3.0 AIRS data currently available from the DAAC show virtually identical biases. The standard deviations of the differences between MLS and AIRS are equal to 58%, 73%, and 53% at 316, 215, and 147 hPa, respectively. Although this scatter is quite large, we see a distinct improvement over v3.0 AIRS, for which these values are 64%, 125% and 88%. The improvement in the scatter from v3.0 to v4.0 AIRS data versions is apparently a consequence of better quality screening flags. Future work is required to better understand the impact of the different horizontal and vertical footprints of these instruments.

B. Ft. Sumner Comparisons

Figure 14 gives results of the Ft. Sumner comparison between the two closest MLS H_2O profiles and the FIRS and MkIV measurements during the September 23/24 balloon flight. Above 100 hPa the agreement between MLS and FIRS is excellent, the largest difference is smaller than ∼0.3 ppmv (or ∼5 %); pwqd somewhat larger differences (but still relatively small) are found with the MkIV profile, the largest difference being ∼0.5 ppmv, or 10 %. At 100 hPa and below, the differences are larger, but the issue in this region of possible MLS profile oscillations and the larger variability make the comparison there more challenging. Differences of

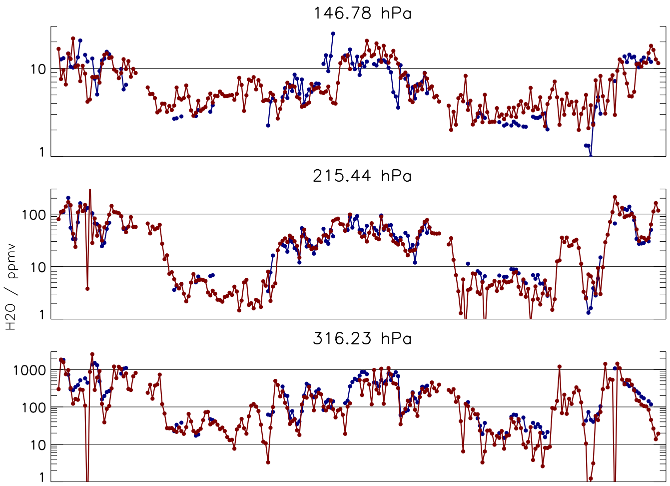

Fig. 13. Typical comparison between MLS (red) and AIRS (blue) retrieved H_2O abundances in the upper troposphere at the pressures indicated, for one orbit on 5 Nov. 2004. Time goes from right to left, and the orbit starts at the equator descending node. According to quality acceptance criteria for each of these datasets, the fraction of rejected profiles along the MLS track is $\sim$ 3% for MLS and 51% for AIRS, based on the two days that we have analyzed so far.

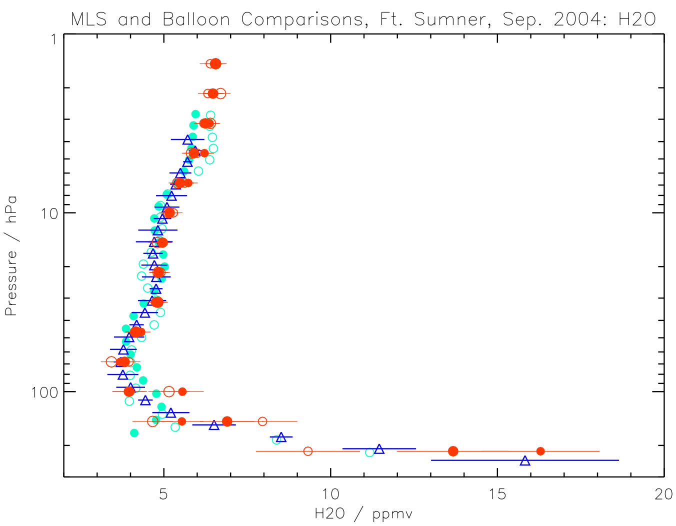

Fig. 14. Similar to Figure 8, but for H_2O Ft. Sumner data versus MLS; MLS profiles (red symbols) are compared to MkIV (blue triangles) and FIRS-2 (cyan) profiles on Sep. 23/24 2004.

up to $\sim$30 % can be seen between MLS and the other 2 instruments, although the precision values and limited statistics preclude one from stating that a clear bias exists between any of these measurements.

V. Hydrogen Chloride

The standard product for version 1.5 HCl is taken from the 640 GHz (Core+R4) retrieval. The recommended vertical range is between 100 and 0.2 hPa. The latter pressure is a conservative limit based on single-profile precisions and the limited influence from *a priori*; averages may enable the use of this product at higher altitudes, but this will require further analyses. The estimated single-profile precision ranges from $\sim$0.1 ppbv (lower stratosphere) to 0.5 ppbv (lower mesosphere), or 5 to 15 %. This is typically close to the scatter based on the retrieved profile variability, evaluated in a narrow latitude band centered around the equator, where atmospheric variability is expected to be small, except at the top end of the profile, where the scatter tends to be somewhat smaller than the estimated precision (at 0.2 hPa, a scatter of 0.35 ppbv is typical). The vertical resolution for HCl is $\sim$3 km in the lower stratosphere, and degrades to 5–6 km near 1 hPa and 7 km at the top recommended level of 0.2 hPa.

Simulations indicate excellent closure (in comparisons of retrieved and simulated 'truth' profiles) for HCl in the stratosphere and lower mesosphere, typically to better than $\sim$ 1 %. The simulations also show that systematic biases tend to increase to 0.1 ppbv or more at the lowermost pressure (147 hPa) used in the retrievals for this product; this can amount to more than 30%, for the small abundances often found at this altitude. This, and the fact that the random error also increases significantly, especially in percent, at this lower end of the profiles, leads us to be cautious about the usefulness of the current MLS retrievals below 100 hPa, except possibly at high latitudes, where larger HCl abundances can more often be found.

A. Global Comparisons

In a manner similar to the ozone comparisons, we provide in Figure 15 average results for the MLS and HALOE HCl profiles during the January through March 2005 time period. This plot indicates that, on average, MLS HCl is typically high relative to HALOE by about 0.2 to 0.4 ppbv ($\sim$10 to 15%). This is in contrast to the comparison of MLS and ACE HCl abundances for roughly the same time period and over some similar latitudes (even if not exactly at the same place and time), as seen in Figure 16. The MLS HCl values are typically within $\sim$5 % of the ACE values, certainly in the upper stratosphere and in the more quiescent SH lower stratosphere. The larger differences in the NH are probably associated with the more disturbed conditions of NH high latitude winter; These results agree overall with those of [33], who quote that ACE HCl (version 1.0) abundances are 10 to 20 % larger than those from HALOE, based on a more limited sampling of 32 coincident profiles, mostly in July 2004. We have observed a similar behavior between HALOE, MLS, and ACE zonal means from August and September 2004 observations at low and high latitudes (not shown here). The exact cause of the disagreement with HALOE is not known at this time, but there were early indications that HALOE measurements of HCl were on the 'low side' of other observations, by about 15%, as mentioned in [37]; at that time, the statistical significance of the differences was not readily apparent, given the smaller number of comparisons versus Atmospheric Trace Molecule Spectroscopy (ATMOS) and balloon-borne profiles.

Despite the apparent bias issue for HCl, which requires further investigation, we find that latitudinal variations for this product agree well between HALOE and MLS (from plots similar to Figure 2, not shown here); also, a systematic bias should have little impact on chlorine trend information

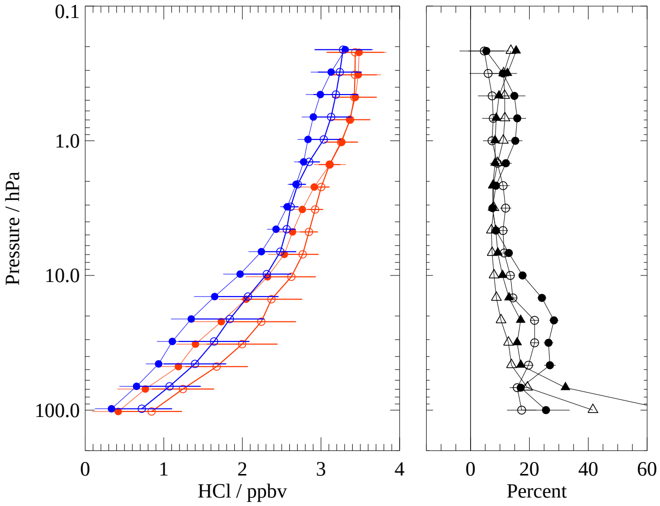

Fig. 15. As Figure 4, but for MLS and HALOE HCl comparisons. A total of 329 matched profiles was used in this case.

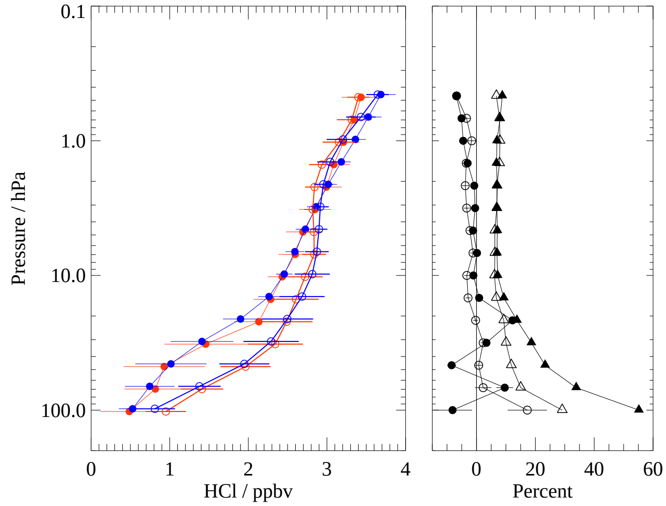

Fig. 16. As Figure 3, but for MLS and ACE HCl comparisons. A total of 623 matched profiles was used in this case.

obtained by HALOE so far (see [38], [39]), as long as the potential error source is time-invariant. There are implications if values of total chlorine in the stratosphere are indeed as large as indicated by MLS or ACE, and it might seem that global MLS observations of 3.5 ppbv HCl near 0.2 to 0.5 hPa are high, since they could imply about 3.7 ppbv of total chlorine, with $\sim$0.3 ppbv uncertainty as a ‘two sigma’ preliminary estimate of the MLS accuracy. Expectations based on ground-based source gas abundances and subsequent transport into the upper stratosphere may come in closer to 3.4 ppbv (based on [40] and *S. Montzka, private communication, 2004*). The uncertainty in this number requires further study, even if it may seem that it should not easily exceed 0.1 ppbv. Any difference in total chlorine abundance between the surface and 50 km should be explainable by a combination of (small) errors in the tropospheric total chlorine, in the effective time lag for transport of chlorine from the surface to 50 km, and in the HCl data from MLS (and ACE) in this region. This requires some further analyses.

B. Ft. Sumner Comparisons

Figure 17 gives results of the Ft. Sumner comparison between MLS HCl profiles and both the OMS profiles of ALIAS–II HCl, on September 17, 2004, and the remote MkIV and FIRS measurements from the September 23/24 BOS flight. There is generally good agreement between MLS and these balloon datasets, given the precision values for individual profiles. There are some altitudes where the MkIV and FIRS-2 measurements appear to differ by about 10 %, although this is not inconsistent with their combined errors. Near 3 to 5 hPa, MLS values tend to be about 10% higher than the infrared balloon values, but not in a statistically significant way. The ALIAS–II *in situ* HCl values are higher than the remote infrared retrievals of a week later, and there are indications of a similar increase in the MLS profiles also, between 10 to 30 hPa.

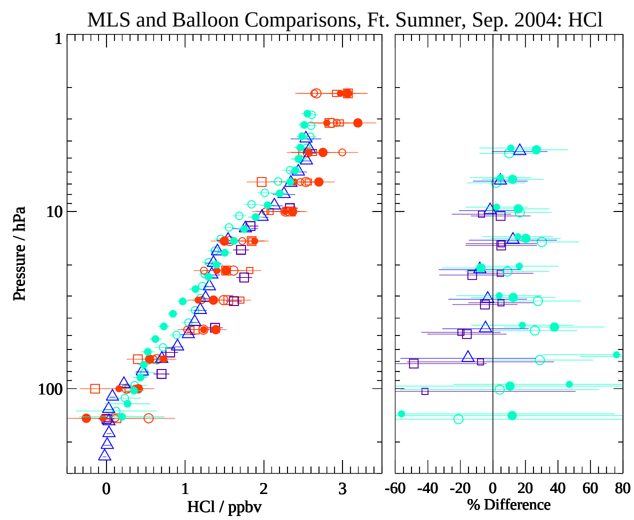

Fig. 17. Similar to Figure 8, but for HCl data. Left Panel: this compares MLS (red symbols) to MkIV (blue triangles) and FIRS-2 (cyan) profiles on Sep. 23/24 2004. Also shown (purple squares) is the ALIAS–II Sep. 17 (*in situ*) HCl profile retrieval, to be compared to the MLS values (red squares) for that day. Right Panel: Percent differences (for MLS minus balloon data) are shown, with symbols referring to the balloon measurements mentioned in the left panel caption. Error bars give twice the random error in these differences.

VI. NITROUS OXIDE

The standard product for N_2O in version 1.5 is derived from the 640 GHz (Core+R4) observations. The v1.5 N_2O data are considered useful for scientific study from 100 hPa up to 1 hPa, though systematic errors (particularly in the lower stratosphere) remain to be investigated. The Level 2 software reports an estimated precision for N_2O of about 15 ppbv from $\sim$22 – 2.2 hPa, worsening above and below to about 30 ppbv at 100 and 1 hPa. The scatter observed in the data from 10°S to 10°N agrees well with this estimate in the mid- and upper-stratosphere, and indicates that the precision in the lowermost stratosphere may be closer to 15 ppbv rather than the reported

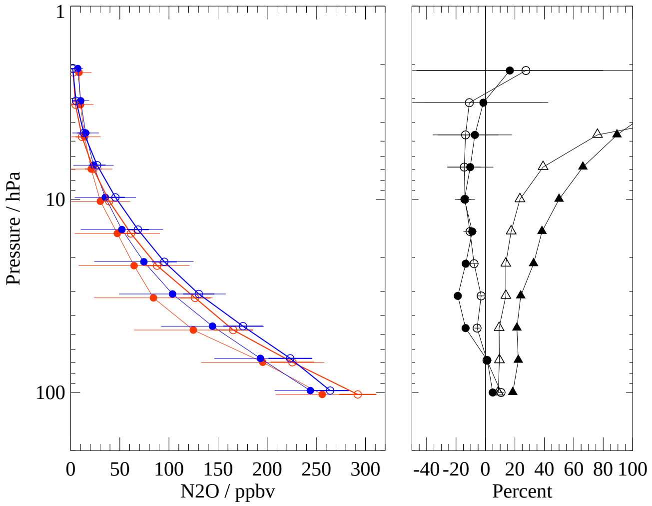

Fig. 18. As Figure 3 but for MLS and ACE N_2O comparisons. A total of 622 matched profiles was used in this case.

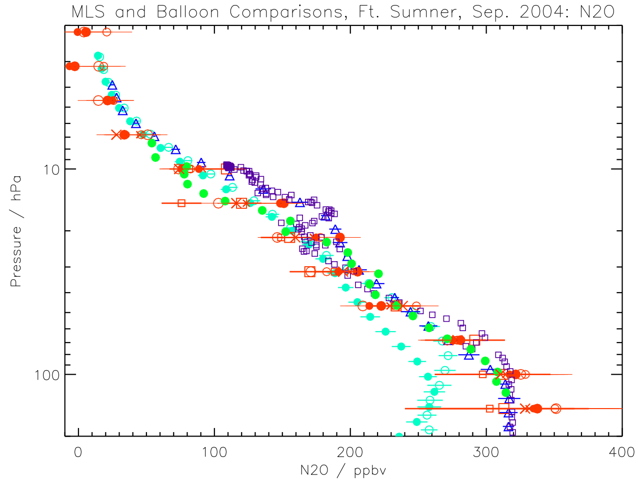

Fig. 19. Similar to Figure 8, but for N_2O Ft. Sumner data versus MLS; MLS profiles (red symbols) are compared to MkIV (blue triangles) and FIRS-2 (cyan) profiles on Sep. 23/24 2004. Also shown is the LACE Sep. 17 (*in situ*) N_2O profile (purple squares), to be compared to the MLS values (red squares) for that day, and the CWAS profile (green dots) of Sep. 29 (*in situ*), to be compared to the MLS values (red crosses) for that day. Error bars are not shown for LACE or CWAS but these should be quite small (less than 1 to 2 % total error).

30 ppbv. The vertical resolution of N_2O is ~4 km through most of the stratosphere, worsening to ~5 km in the region below 46 hPa. Simulation studies indicate that biases of order 60 ppbv (20%) are possible in the lower stratosphere polar vortex regions. These are due to approximations made in the forward model to increase data processing speed. Also, occasional high biases or order 30% have been observed at 100 and 68 hPa, possibly indicating a slight instability in the retrieval that will be investigated further as part of the development of future versions.

A. Global Comparisons

Figure 18 summarizes matched comparisons of MLS N_2O with those of the ACE instrument. Generally good agreement is seen between the instruments, with mean biases typically less than 20%, and the standard deviation of the differences between the observations being around 40% in the lower stratosphere, increasing (as expected due to decreasing N_2O abundances) in the upper stratosphere.

B. Ft. Sumner Comparisons

Figure 19 shows the comparison between MLS N_2O profiles and the remote measurements from the FIRS and MarkIV instruments September 23/24 2004 Ft. Sumner balloon flight described in the Introduction and ozone sections. The comparisons are very encouraging with MLS N_2O agreeing with the balloon instruments to the level one would expect from the precision estimated on the MLS data. This implies we can be confident that any biases are within the combined precision reported by the instruments of around ±15 ppbv (5% in the lower stratosphere, increasing to 30% around 2.2 hPa). MLS data cannot easily track the apparent difference observed in *in situ* data between September 17 and September 29, but we expect that averaging a number of nearby MLS profiles would enable detection of such variations, if they occur on a sufficiently large scale. MLS observations in the lower stratosphere seem to be more consistent with those of the Mark-IV and *in situ* data than the FIRS data, which seem to be on the low side of the other balloon data there.

VII. Nitric Acid

The standard product for HNO_3 in version 1.5 is derived from the 240 GHz (Core+R3) observations at and below 10 hPa, and from the 190 GHz (Core+R2) observations at and above 6.8 hPa. The v1.5 HNO_3 data are considered useful for scientific studies from 147 to 3.2 hPa; results from simulations indicate that large systematic biases limit the scientific usefulness of the HNO_3 retrievals outside of this range. Over most of the recommended vertical range, the estimated single-profile precision reported by the Level 2 software varies from ~1.0 to 1.5 ppbv; the observed scatter in the data, evaluated in a narrow latitude band centered around the equator where atmospheric variability is expected to be small, suggests a measurement precision of ~1 ppbv throughout the profile. The vertical resolution of HNO_3 is ~3.5 km over the range 100 to 10 hPa, degrading to ~4.5 km at 3.2 hPa.

Simulations indicate that average biases are small over the range 147–3.2 hPa, with an overall accuracy of better than 10%. In contrast to the simulations, however, preliminary comparisons with a climatology based on seven years of UARS MLS measurements [41] suggest that EOS MLS HNO_3 may be biased high by several ppbv near the profile peak. Much closer agreement with the UARS climatology is generally found at other latitudes/altitudes/seasons. The apparent high bias in the peak mixing ratios, also evident in other comparisons as discussed below, will be explored in more detail in future studies. In addition to more 'traditional' types of analyses, we note that probability density function (PDF) analyses can also point to biases between datasets. For

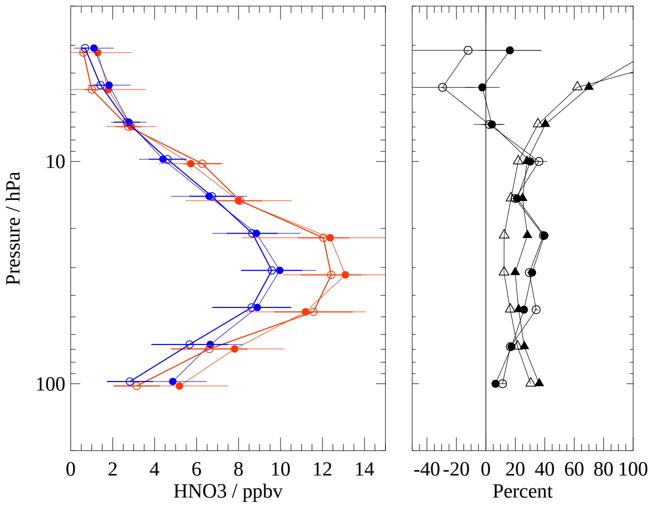

Fig. 20. As Figure 3, but for MLS and ACE HNO_3 comparisons. A total of 621 matched profiles was used in this case.

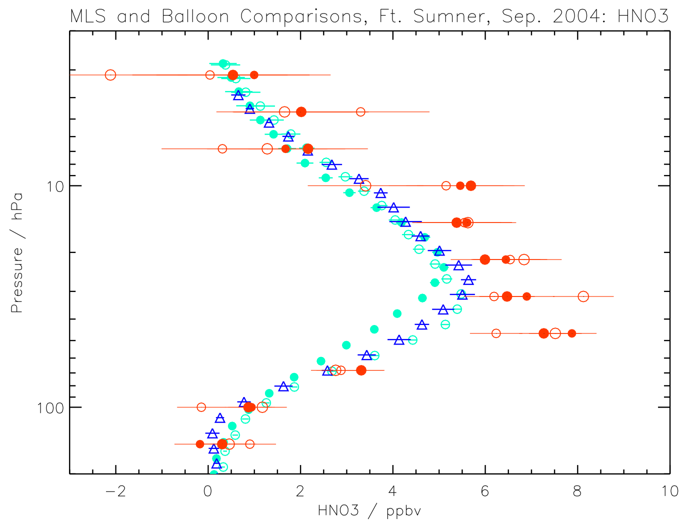

Fig. 21. Similar to Figure 8, but for HNO_3 Ft. Sumner data versus MLS; MLS profiles (red symbols) are compared to MkIV (blue triangles) and FIRS-2 (cyan) profiles on Sep. 23/24 2004.

example, this approach (see [42]) shows that the UARS MLS HNO_3 abundances are significantly larger than those obtained by the Cryogenic Limb Array Etalon Spectrometer (CLAES) infrared measurements, as shown previously by [43].

A. Global Comparisons

In a manner similar to the ozone comparisons, we provide in Figure 20 average results for the MLS and ACE HNO_3 profiles during the January through March 2005 time period, at mostly middle and high latitudes. This plot indicates that, in an average sense, MLS HNO_3 is high relative to ACE by 2–3 ppbv ($\sim$ 30%) at the levels surrounding the profile peak. Average agreement between the two satellite measurements is much better (typically within $\sim$10%) near the top and bottom of the profile. Despite the apparent offset between MLS and ACE near the profile peak, however, comparisons of nearly-coincident individual measurements (not shown) show good agreement in capturing the overall shapes of the HNO_3 profiles and tracking variations in them.

B. Ft. Sumner Comparisons

Figure 21 shows results of the Ft. Sumner comparison between MLS HNO_3 profiles and those of MkIV during sunset on Sep. 23, and FIRS-2 on September 23 (daytime Aura overpass) and 24 (nighttime Aura overpass). Again, MLS mixing ratios can exceed those measured by the balloon instruments by as much as 3 ppbv at the levels around the profile peak, with the magnitude of the discrepancy well outside the combined error bars in some cases. As in the previous comparisons, agreement is typically much better away from the profile peak at the top and bottom of the vertical range.

VIII. Carbon Monoxide

The standard product for CO in Version 1.5 comes from the 240 GHz (Core+R3) observations, using the emission line at 230 GHz. The useful vertical range of the current retrievals is 215 to 0.0022 hPa, although some artifacts are to be noted (see below). The single-profile precision ranges from 20 ppbv between 215 and 22 hPa, then increases approximately inversely with pressure to reach 1 ppmv at 0.0022 hPa. The vertical resolution of CO is $\sim$4.5 km up to 0.1 hPa, $\sim$6 km above 0.1 hPa.

A. Global Comparisons

In a manner similar to the ozone comparisons, we provide average results for the MLS and ACE CO profiles during the 2005 January through March time period. ACE CO observations have been described recently (see [44]). Figure 22 gives results of the matched comparisons. In both the ACE and MLS data, there is marked north-south asymmetry. Most of the profiles are from high latitudes in each hemisphere and there is strong descent of mesospheric air into the polar stratosphere during the winter, giving the increased mixing ratios seen in the northern hemisphere curves. The ACE and MLS data have the same *general* behaviour, but there are several artifacts in the MLS data:

- From 10 hPa upwards, there are strong oscillations. These are most obvious in the southern hemisphere, but the oscillations of the same magnitude occur in the northern hemisphere; the log scale compresses them in the figure. The oscillations are about 3 times as large as the estimated precision and are thought to be a result of insufficient smoothing in the retrievals.
- The CO retrieval at 68–32 hPa at high latitudes appears to be affected by the large mixing ratios of HNO_3 found in these regions: there is a correlation between the fields, seen in maps and in scatter plots (not shown). There does not appear to be any dynamical or chemical reason for these correlations and there are weak HNO_3 lines in the frequency band used to measure CO. It is not yet certain if the forward model needs to be improved, or if the signals from the two molecules cannot be distinguished in these conditions.

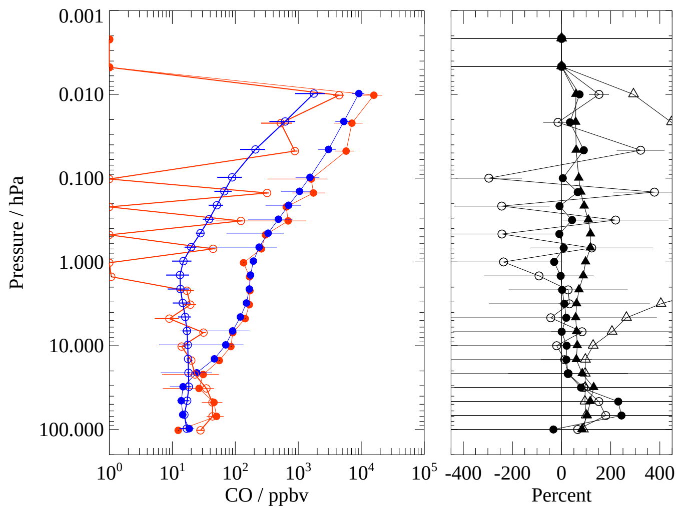

Fig. 22. As Figure 3 but for MLS and ACE CO comparisons, and with a log axis for mixing ratio. A total of 616 matched profiles was used in this case. In the left-hand panel, mixing ratios less than 1 ppbv (or negative) are set to 1 ppbv to better display the oscillation in the average MLS profiles.

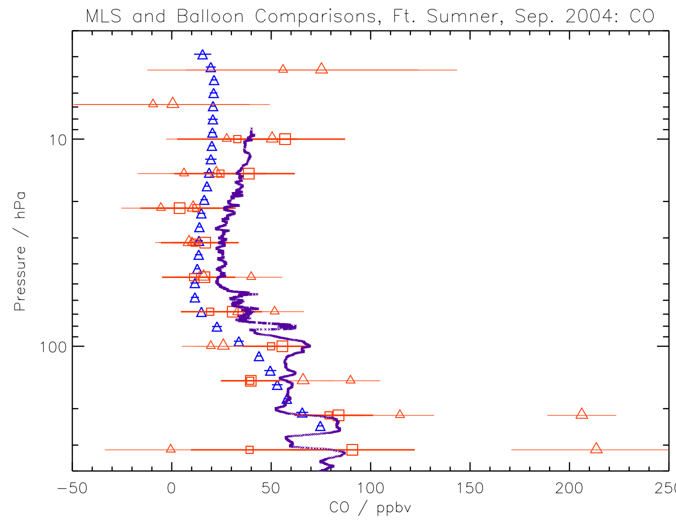

Fig. 23. Similar to Figure 8, but for CO data; this compares MLS (red symbols) to MkIV (blue triangles) on Sep. 23 2004. Also shown (fine resolution line of purple crosses) is the ALIAS–II Sep. 17 (*in situ*) CO profile retrieval, to be compared to the MLS values (red squares) for that day. Note the possible high bias in ALIAS-II stratospheric values versus MkIV data; this may be caused by contamination of ALIAS-II data, as mentioned in the Introduction

In the 22–0.22 hPa range for the northern hemisphere, where the MLS profiles are not strongly affected by oscillations or nitric acid, MLS has a $< +40$ % bias compared to ACE, reducing to $+5$ % in the middle stratosphere. Smoothing the retrievals for the southern hemisphere should give similar biases. In the upper mesosphere and lower thermosphere, MLS has a 50–100% positive bias compared to ACE.

B. Ft. Sumner Comparisons

Figure 23 gives results of the Ft. Sumner comparison between MLS CO profiles and both the *in situ* profiles of ALIAS–II, on September 17, 2004, and the remote MarkIV measurements during the sunset of September 23, about a week later. The comparisons with the balloon instruments cover the upper troposphere and lower/middle stratosphere. The oscillations in the MLS data seen in Figure 22 are also seen starting at the uppermost 3 MLS levels in Figure 23. The general behaviour is similar between MLS and the balloon instruments and, in this small sample, consistent within the errors. MLS has a positive bias at 215 hPa and the 316 hPa has large scatter. MLS generally retrieves small or negative mixing ratios near 32 hPa in the tropics/sub-tropics; this can be seen in Figure 23

IX. Summary and Future Plans

These early comparisons between MLS version 1.5 data and other satellite and balloon-borne measurements reveal good overall agreement in the stratosphere, with some average differences as low as 5 to 10 %, for the January–March 2005 time period. This is particularly encouraging, when one considers that we have not yet completely optimized our comparison methods, and that most 'established datasets' do not often agree with each other to better than the 5 % level. However, tracking changes in the atmosphere is often more important than optimum accuracy for absolute values. In most instances, we have observed that the variability of the matched profiles between MLS and other correlative (global) datasets is well correlated, and that variations between individual profiles (not shown in detail here) exhibit very similar behavior, for example in and out of the polar vortex. This, and the latitudinal changes that we have observed in the various datasets, indicates that the retrieved MLS profiles are indeed tracking changes that are very consistent with those observed by other instruments; there is also very good consistency in the retrieved MLS fields with potential vorticity (see [4]).

Some MLS retrieval issues we are aware of, not all of which are shown or discussed in detail here, include artifacts that are apparent without invoking correlative datasets, such as oscillations or (typically small) negative average abundances at some heights. We intend to address these issues in a next retrieval version. As mentioned by [6], plans for the next MLS software version and public dataset will also include work on a higher vertical resolution product for water vapor, as well as a faster forward model. The latter improvement, and potential revisions in spectroscopic parameters, should allow for more accurate calculations, especially under non-linear conditions, and for probing deeper into the troposphere. Updated calibration results may also provide some improvements.

Version 1.5 MLS temperature comparisons with other global datasets indicate that MLS stratospheric temperatures have a 1–2 K warm bias. It is anticipated that future MLS data versions will utilize radiances from the 190 and 240 GHz radiometers to improve vertical resolution in the upper troposphere and stratosphere and to possibly reduce this bias.

For O_3, the comparisons so far indicate overall agreement at roughly the 5–10 % level with stratospheric profiles from SAGE II, HALOE, POAM III, and ACE. Atmospheric variability is generally represented in a similar way by MLS and these other satellite observations, when comparing coincident profiles. In comparison to the different occultation datasets investigated so far, MLS ozone tends to be slightly larger (by varying amounts) in the lower stratosphere, and slightly smaller in the upper stratosphere. The standard MLS product leads to a better overall match in the stratosphere than do the O_3 products from the other MLS radiometers, except in the lower mesosphere, where somewhat of a high bias is evident in the standard MLS product. In the mesosphere, diurnal changes and other issues will require more careful investigations; although not shown here, daytime MLS coincidences with occultation profiles produce a better fit than do the nighttime MLS coincidences. In the near-term, MLS validation studies will add more emphasis on the important regions of the upper troposphere and lower stratosphere (UT/LS), especially in the tropics.

For H_2O, the comparisons so far broadly indicate that MLS H_2O can be trusted at the roughly 10 % level of accuracy; however, some artificial oscillations tend to be present in the lower stratospheric portion of many MLS profiles, something to be addressed in a future software version. Besides doing more of these types of comparisons, future plans include analyses versus frost-point sondes and radiosondes, especially for the interesting tropical regions. Aircraft campaign studies are in progress to help address systematic differences between *in situ* measurements, an area that needs improvement in order to further assist in the validation of satellite measurements.

For HCl, we find that latitudinal mean variations and variability compare well with those from HALOE, but the MLS values are systematically larger than the HALOE abundances by 0.2 to 0.4 ppbv. This amounts to 10–15 % in the upper stratosphere and lower mesosphere, where HCl is a measure of total chlorine. The HALOE values have been shown to be smaller than ACE HCl abundances by a similar amount, and the ACE and MLS comparisons shown here indicate excellent agreement (within a few %) in the upper stratosphere, with only slightly larger percent differences in the lower stratosphere. While it may well be that HALOE underestimates the absolute values of HCl and total chlorine, more work is needed to understand how to best reconcile the MLS and ACE 50-60 km HCl abundances with total surface chlorine estimates, given the various error sources. The Ft. Sumner comparisons of MLS HCl versus measurements from ALIAS, MkIV, and FIRS-2 indicate overall agreement, within the combined likely errors. Of future interest will be more studies of MLS HCl and correlative data as a function of latitude and time.

For N_2O, comparisons so far are very encouraging, indicating agreement at the 20 % level. Occasional biases of order +30% in the lowermost stratosphere remain to be investigated, and will be addressed in the next software version.

MLS HNO_3 data are useful for scientific studies from 147 to 3.2 hPa. On the basis of comparisons with nearly-coincident satellite (ACE) and balloon-borne (ALIAS and MkIV) measurements, the MLS HNO_3 retrievals appear to be biased high by about 3 ppbv ($\sim$30%) at the levels surrounding the profile peak. Much better agreement is seen near the top and bottom of the vertical range, and the overall shapes of the profiles and the variations in them are captured well in the MLS data. The altitude, latitude, and seasonal dependence of the apparent high bias in the MLS v1.5 HNO_3 data will be explored in more detail in future validation studies, both through analysis of potential shortcomings in the MLS retrieval system and comparisons with additional correlative (ground-based, aircraft, balloon, and satellite) data sources.

For CO, the comparisons so far indicate that (with smoothing) MLS overestimates stratospheric CO by $\sim$40%. There are strong oscillations in the upper stratosphere and mesosphere, and CO observations in the polar lower stratosphere are affected by HNO_3. In terms of future plans, comparisons will continue with both ACE profiles and the Odin Sub-Millimetre Radiometer dataset. In the upper troposphere and lower stratosphere, various aircraft campaigns (see below) will provide coincident measurements, and comparisons will be made with the TES observations, as well as with data from the Measurement Of Pollution In The Troposphere (MOPITT) experiment, and aircraft data from commercial flights participating in programs such as the Measurement of OZone and wAter vapour by Airbus In-Service airCraft (MOZAIC).

The coming years will see much continuing work on both the MLS retrievals and validation efforts such as those described herein; most of the results shown here do not address MLS retrievals at low latitudes, an important topic for further study. Analyses of aircraft and sonde data from previous 'Aura Validation Experiment' (AVE) campaigns are on-going and will be discussed elsewhere; this includes the WB-57 measurements made in October/November 2004 from the Houston area, and the DC-8 measurements from the Polar Aura Validation Experiment, PAVE, based in New Hampshire, exploring high latitudes during February/March 2005. Another Houston-based campaign is planned for June 2005, along with tropical O_3 and H_2O sonde data from Costa Rica. Future aircraft and ground-based campaigns of interest to MLS should also include more upper tropospheric measurements of O_3 and CO, under polluted conditions. The Intercontinental Chemical Transport Experiment (INTEX) campaign planned for the spring of 2006 from the West Coast of the United States should address some of these goals. High latitude balloon measurements could help to further cross-validate Aura and ACE (and other) measurements. Future validation work will include cross-platform comparisons for the various Aura instruments, and will also shift towards more seasonal and longer-term comparisons, as well as to less 'traditional' validation studies (e.g., comparisons involving air masses with the same potential vorticity, or using PDFs, as in [42]) .

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